Analysis of room-temperature results on normally conducting and superconducting channels through polymer films

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ABSTRACT

There are strong reasons from dc and pulsed-current measurements, and from thermal conductivity results, for thinking that narrow channels through films of oxidised atatic polypropylene (OAPP) are superconducting at room temperature. It is thought that the conducting channels, with diameters less than or of the order of a micrometre, are composed of smaller nanofilaments, with diameters of the order of a nanometre. In the present paper a possible explanation is given of measurements which show that the average resistance of non-superconducting channels through films increases with film thickness more slowly than linearly. This result is interpreted in terms of how the Bose condensation temperatures of bosons in arrays of nanofilaments depend on the length and numbers of filaments, and examples are given of parameters of the arrays which could explain the data. The dispersion for the bosons is assumed to consist of a sum of linear and quadratic terms, which is an approximate type of dispersion reported for Cooper pairs. In order to fit the data with the model used, it is necessary to suppose that values of superconducting T_c for channels composed of large numbers of filaments are only slightly above room temperature. It is argued that larger T_c's reported in 1989 when currents of 0.5 A are passed through channels may arise because (a) currents concentrate in a subchannel of smaller width than the original channel, and (b) current-current interactions draw the filaments of the subchannel sufficiently close together to increase the transverse bandwidth and T_c in the model by the required amount.

§1. INTRODUCTION

The first indication that narrow channels through films of oxidised atactic polypropylene (OAPP) might be superconducting at room temperature came from a finding from resistance measurements with microprobes a few μm in diameter at various points on the top surface of films of OAPP on a conducting substrate. Some points showed very low resistance independent of the film thickness and about equal to that of a contact put directly on the substrate (see

figure 1, taken from figure 17 of Grigorov and Smirnova 1988). (See also Enikolopyan *et al.* 1989, Arkhangorodskiĭ *et al.* 1989).

More convincing evidence came in 1990 and 1991 from three types of results:
(i) Estimates of lower limits on conductivity for channels several orders of magnitude greater than that of copper by direct (Arkhangorodskiĭ *et al.* 1990) and indirect (Demicheva *et al.* 1990) methods;
(ii) Destruction of superconductivity by non thermal means by pulsed currents, with critical current densities of the order of 5×10^9 A cm^{-2} (Demicheva *et al.* 1990); and
(iii) The finding of negligible electronic thermal conductivity in the channels, violating the Wiedemann-Franz law by several orders of magnitude (Grigorov *et al.* 1991).

Some unusual magnetic effects (Enikolopyan *et al.* 1989, Grigorov *et al.* 1993, Rogachev and Grigorov 2000) involving large diamagnetism have been seen, and are probably associated with channels which form closed loops. A possible interpretation of large diamagnetism in low magnetic fields at room temperature in some samples has been given recently by the present author (Eagles 2002). Other magnetic effects such as superparamagnetism and metamagnetism (Smirnova *et al.* 1988, Enikolopyan *et al.* 1989, Grigorov *et al.* 1996, Rogachev and Grigorov 2000) are thought to be associated with conducting channels which do not form closed loops.

The channels have maximum diameters of the order of 1 μm (Demicheva *et al.* 1990), but for theoretical reasons (Grigorov 1990, 1991, Grigorov *et al.* 1990) it is thought that the channels are composed of large numbers of smaller nanofilaments, known as superpolarons, of diameters of the order of one to a few nm. The only two types of published theory for superconductivity in the channels of which the author is aware (Eagles 1994a, 1994b, Grigorov 1998) make use of the assumption that the channels are composed of many quasi one-dimensional filaments. The present author (Eagles 1998a) and Grigorov and coworkers (see e.g. Grigorov and Rogachev 2000) have very different suggestions as to the origin of the magnetism in the non-closed-loop channels, although both depend on the above assumption.

§2. SUPERCONDUCTING AND NORMALLY CONDUCTING CHANNELS

When the resistance is measured between a conducting substrate and microprobes of diameters of approximately 10 μm placed on top of OAPP films, distribution functions of the resistance for two different thicknesses of film and for contacts put directly on the substrate are shown in figure 1, taken from Grigorov and Smirnova (1988).

Note that for the films there are three types of points (i) insulating, (ii) mediumly conducting, and (iii) highly conducting. For the mediumly conducting points, the lowest resistance for the 50 μm film is slightly more than 30 times as large as the lowest resistance for the 0.3 μm film, implying that the cross section of channels associated with these points is about five times larger than for the 50 μm film if the resistivity is the same for the material of these channels in both films. In a later paper (Enikolopyan *et al.* 1989), the departure from linearity is smaller, and, for the results presented there, we can deduce that the cross section of channels with the lowest normal resistance in the thinner films is only about twice as large as the corresponding cross section for the thinner films. Later in the paper we use parameters which give a value of four for this ratio, in between the 1988 and 1989 results.

The channels associated with the highly conducting points are probably superconducting (see the Introduction), and in this paper we explore the hypothesis that the reason for the apparently larger cross section of the normally conducting channels with the lowest normal resistance in the thicker films is that a larger cross section is required for superconductivity to occur at a given temperature in channels of larger lengths. The specific model which we use
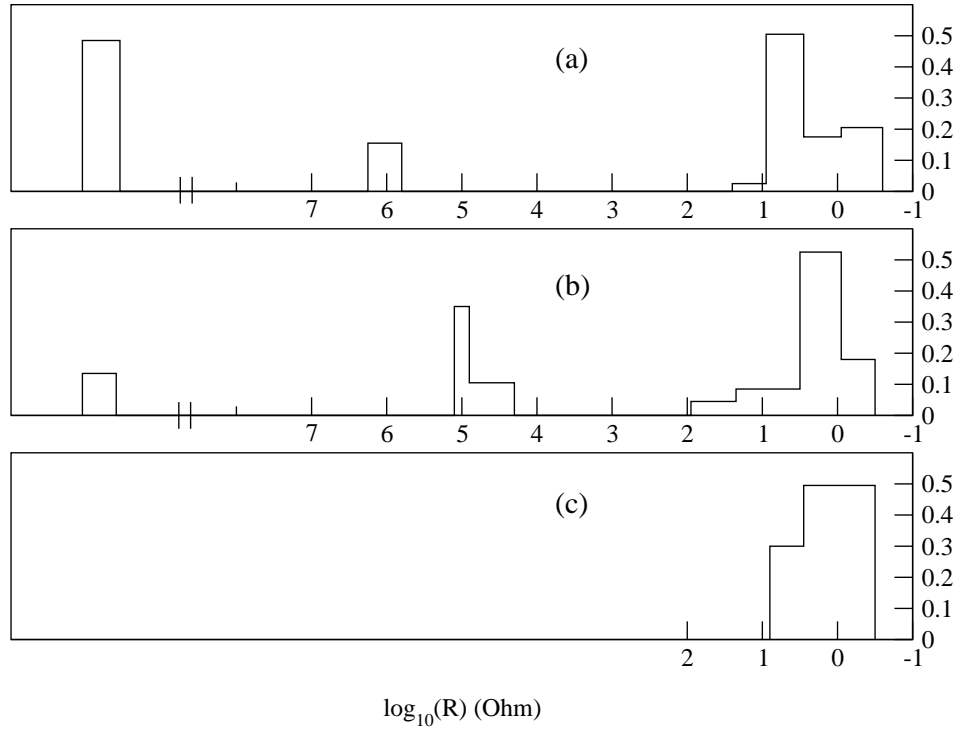
Figure 1: Distribution function for resistance R at room temperature from microprobes to a metallic substrate: (a) For a sample of oxidised atactic polypropylene (OAPP) of thickness 50 μm, (b) for OAPP with thickness 0.3 μm, and (c) for the metallic substrate without any polymer film. [Reproduced approximately from figure 17 of Grigorov and Smirnova (1988)].

to examine this hypothesis is a model of charged bosons on an array of quasi-one dimensional filaments, with a boson dispersion consisting of a combination of linear and quadratic terms, which is found to be a fair approximation for the dispersion of Cooper pairs (Adhikari et al 2000). Although a Cooper-pair model gives unrealistically small binding energies for large Fermi energies or for weak coupling because of neglect of what happens below the Fermi surface, we think it may be more realistic when pair binding energies are large compared with the Fermi energy as in that case what happens below the Fermi energy is less important. Even for the case of large Fermi energies, although the Cooper-pair binding energies are too small, it is possible that the Cooper-pair model may give the correct form of the wave-vector dependence of pair binding energies relative to those for zero wave vector.

Our calculated values of T_B will be close to superconducting transition temperatures T_c if and only if the pairs do not break up at a temperature lower than T_B, i.e. if we are on the Bose-gas side of the BCS-Bose gas transition. This transition was first studied by Eagles (1969). In the BCS theory the pair binding energy is $3.5k_BT_c$, and is somewhat larger in its strong-coupling extensions. Thus a necessary condition for our theory to be valid is that the binding energy of pairs should be larger than $3.5k_BT_B$, where T_B is calculated neglecting pair break-up.

We also mention that, for arrays with small numbers of nanofilaments, the resistance at the condensation temperature cannot be expected to be zero, because of either thermal (Langer and Ambegoakar 1967, McCumber and Halperin 1970) or quantum (Zaikin *et al.* 1997, Zhao 2003) phase slips of the order parameter. The ultra-high conductivity of several orders of magnitude larger than that of copper at room-temperature mentioned in the Introduction has only been established for relatively large arrays of nanofilaments forming channels of diameter of the order of a micrometer, for which effects of phase slips are expected to be negligible. For smaller arrays the only experimental lower limits on the conductivity are much lower and only approximate, based on the lack of noticeable length dependence of the resistance of the channel plus contact resistance of the highly conducting channels as indicated in figure 1.

§3. CONDENSATION OF BOSONS IN AN ARRAY OF NANOFILAMENTS

In this section we use an idealised model of conducting channels composed of non-interacting bosons on a square array of quasi one-dimensional filaments of length L and transverse dimension D measured from point half a filament separation a_T beyond the centers of the edge filament rows. The filaments are assumed to be sufficiently narrow that the bosons in a filament can be assumed to be in the lowest possible state with respect to motion perpendicular to the filaments at all temperatures of interest. We suppose that boson wave functions vanish at the edges of the array, and neglect any periodicity in the potential along the length of the array. Then we suppose that the boson states in the array can be characterised by the magnitudes of wave numbers K_1, K_2 and K_3 in the two transverse and longitudinal directions, where K_1 and K_2 take on discrete values ranging from π/D to $n_T(\pi/D)$, and K_3 takes on values of $k\pi/L$, where k runs from 1 to ∞. For computational purposes we approximate the sum to infinity by a finite sum with a suitably high upper limit.

Based on the fact that the dispersion relation for Cooper pairs can be approximated as a combination of linear and quadratic terms (Adhikari *et al.* 2000) we assume that the boson energies $E_B(K_1, K_2, K_3)$ measured from the lowest state can be written as

$$E_B(K_1, K_2, K_3) = \quad A_L[(\pi/L)^2(k^2 - 1) + (d_L/a_T)(\pi/L)(k - 1)]$$
$$+ [W_T/(1 + s_T)][(i^2 + j^2 - 2)/n_T^2 + s_T(i + j - 2)/n_T]. \tag{1}$$

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Here

$$A_L = \hbar^2/2M_L, \tag{2}$$

where M_L is the boson mass in the longitudinal direction, a_T is the interfilament separation, d_L is a dimensionless constant characterising the ratio of the linear to the quadratic terms in the longitudinal boson dispersion relation, s_T is the ratio of the contributions of the linear and quadratic terms in the dispersion to the transverse bandwidth W_T (in a direction parallel to one of the sides of the cross section of the channel), and

$$p = M_L/M_T, \tag{3}$$

where M_T is the boson mass in the transverse direction. W_T, s_T, A_L, p and a_T are related by

$$W_T/(1 + s_T) = pA_L(\pi/a_T)^2. \tag{4}$$

We measure W_T from hypothetical levels with $i = j = 0$ in order to avoid having a dependence of W_T on n_T. We will discuss values of the parameters d_L and s_T in the next section.

The case of zero s_T and d_L (pure quadratic dispersion) was discussed by Eagles (1998b), but with the difference from the case assumed here that a discrete lattice was considered in the longitudinal as well as transverse directions. However, we have recently found, based on work of Alekseev (2002), that for some of the arrays considered there it is difficult to unambiguously define a Bose condensation temperature because of the gradual way in which occupation numbers of the ground state change with temperature. This problem also arises for the smallest arrays for a linear dispersion relation (Alekseev 2001), but for this case the width of the transition is proportional to $1/N_B^{\frac{1}{2}}$, where N_B is the total number of bosons, and is less than about 16% of the condensation temperature when $N_B > 150$.

Condensation into the lowest-energy state cannot take place if the sum of the boson occupation factors over all states except the lowest for a chemical potential situated at the energy of the lowest state is greater than the total number of bosons, N_B. Below the temperature at which this sum is equal to N_B, the occupation of the ground state and of states in its neighbourhood will start to become macroscopic, i.e. of the order of N_B, and so the equality of the sum with N_B determines the condensation temperature T_B in this weak sense (Rojas 1997). For an average linear concentration c of bosons per filament, this criterion can be written as

$$\sum_{K_1,K_2,K_3} \frac{1}{\exp[E_B(K_1, K_2, K_3)/k_B T_B] - 1} = cLn_T^2, \tag{5}$$

where the sum over K_1 and K_2 are sums from $i, j = 1$ to n_T of $K_1 = i\pi/D$ and $K_2 = j\pi/D$, and the sum over K_3 is a sum from $k = 1$ to infinity of $K_3 = k\pi/L$, but the ground level $i = j = k = 1$ is excluded.

As mentioned above, the ground-state occupation for small arrays changes gradually, but we shall only present results for total numbers of bosons equal to 150 or more.

§4. A CHOICE OF PARAMETERS WHICH CAN FIT THE DATA

For superpolarons in elastomers with rotatable polar groups, the potential felt by electrons in the superpolaron arises from about three oriented dipoles per electron surrounding the superpolaron string (Grigorov 1990). Even if arranged regularly, the periodic part of the potential due to these surrounding dipoles is expected to be small, and so the use of a continuum model in the direction of the filaments is probably justified.

E(K) curve at T = 0

In one dimension at $T = 0$ the coefficient of the linear term in the Cooper pair energy at a center-of-mass wave vector K is $\hbar^2 k_F/m_s$ (Casas *et al.* 1998), where m_s is the single-particle mass, while in three dimensions it is half of this. The case of an anisotropic three-dimensional system has not been studied yet, and so we have to make some assumptions. For the direction along the channels where the mass is smallest, we suppose that the one-dimensional result holds. Hence, if the pair binding energy is large compared with the Fermi energy, when the coefficient of the quadratic term is approximately $\hbar^2/4m_s$ (Adhikari *et al.* 2000), then the ratio r of the coefficients of the linear to quadratic terms is

$$r = 4k_{FL}, \tag{6}$$

where we have added the suffix L to denote the longitudinal direction. Using the relation

$$k_{FL} = \frac{1}{2}\pi c_F \tag{7}$$

for one dimensional systems, where c_F is the linear density of fermions, and putting $c_F = 2c$, with c the linear concentration of bosons, we find that

$$d_L = 4\pi(ca_T). \tag{8}$$

A proper calculation of the parameter s_T in Eq.(1) would be difficult. In the Appendix we describe two suggestions which may give approximate upper and lower limits for s_T in terms of the ratio $W_T/(A_L K_{MT}^2)$, where K_{MT} is the maximum tranverse wave vector in the first Brillouin zone, i.e.

$$K_{MT} = \pi/a_T. \tag{9}$$

In order to reduce the number of adjustable parameters by one we take s_T for a given W_T and p equal to the geometric mean of the possible upper and lower limits. It turns out that this mean is independent of W_T and p and is given by

$$s_T = 2^{\frac{1}{2}}, \tag{10}$$

implying

$$p = W_T/[(1 + 2^{\frac{1}{2}})A_L K_{MT}^2]. \tag{11}$$

E(K) curve at finite T

As far as we know, no $E(K)$ curves for Cooper pairs at finite temperature have been published. We expect that, when T is greater than the degeneracy temperature, i.e. when $k_B T_B > E_F$, with E_F the Fermi energy, then the linear term in the dispersion will no longer exist. We make the simplest assumption consistent with this, i.e. that the coefficient of both the longitudinal and transverse linear terms decrease by a factor F given by

$$F = 1 - k_B T/E_F \tag{12}$$

if $k_B T < E_F$, and $F = 0$ when $k_B T > E_F$.

We still have four parameters to vary, and the only pieces of data we have to fit are the ratio of the largest cross sections to give normal conductivity at room temperature for channels of lengths 0.3 and 50 μm, and some results on limits of transition temperatures. Thus we cannot

determine all four parameters. We choose $a_T = 2$ nm, $c = 0.5 \times 10^7$ cm^{-1}, and $p = 0.12$. We then determine a value of M_L which can fit the fact that the ratio of the cross sections of the channels with largest normal resistance at room temperature is about four. For the value of c above, Eq.(7) gives $k_{FL} = 0.5\pi \times 10^7$ cm^{-1}. Solution of Eq. (5) for these parameters for channel lengths of 0.3μm and 50μm are shown as a function of n_T^2 for $M_L = 3.3m_e$ in figure 2. Interpolating between values of n_T^2 of 1, 4 and 9 for which calculations have been done, we find that the 0.3μm channels are normal at room temperature for n_T^2 up to 2, and the 50μm channels are normal for n_T^2 up to 8, and so the ratio of the cross sections of the lowest resistance normal channels for the thicker and thinner films is four, in between the values of about five from figure 1 and of about two from Enikolopyan et al. (1989), as discussed in §2.

From figure 1 the lowest resistance of normal channels for the 0.3μm film is about $2 \times 10^4 \Omega$, which according to figure 2 corresponds to two nanofilaments in the channel. With $a_T = 2$ nm, and a channel length of 0.3μm, the resistivity of the channel is $0.53 \times 10^{-4}\Omega$ cm. With a boson density of $ca_T^{-2} = 1.25 \times 10^{20}$ cm^{-3}, we deduce boson mobilities of about 470 cm^2/Vs. With a charge of $2e$ and a mass $M_L = 3.3m_e$, this corresponds to a relaxation time $\tau = 4.4 \times 10^{-13}$s assuming diffusive transport. For a single-particle mass of 1.65 m_e and $k_F = 0.5\pi \times 10^7$ s^{-1}, the velocity due to the linear term in the dispersion is 1.1×10^7cm s^{-1}. For a pure quadratic dispersion and a mass of 3.3 m_e, the thermal velocity at room temperature $(k_B T/M_L)^{\frac{1}{2}}$ would be 0.37×10^7 s^{-1}, and so the total velocity might be expected to be a bit less than 1.5×10^7 cm s^{-1}, corresponding to a mean-free path $l \approx 70$ nm. The circumference of a channel with two nanofilaments is 12 nm, so we may be on the ballistic side of the quasi-ballistic diffusive transition according to statements of Schönenberger *et al.*(1997). If that is the case, we may deduce that the conduction in the channel may be quasiballistic. In that case the resistance R can be expected to be given by $R = (R_Q/N_p)(L/l)$, where $R_Q = 12.9$ kΩ is the quantum of resistance, N_p is the number of paths through the channel (we use paths here instead of the usual channels as we have already used channel in a different sense), l is the mean-free path, and L is the channel length. Taking $N_p = 2$, this expression implies $l = 100$ nm for $R = 2 \times 10^4 \Omega$ and $L = 0.3\mu$m, not much different from the value given by the diffusive transport calculation.

If there is any periodic part of the potential due to dipoles surrounding the superpolaron, it is probably fairly weak, and so the bare mass of the electrons in the superpolaron string is expected to be close to the free-electron mass. However, we expect that the pairing of electrons in the superpolaron occurs by mediation of very high energy phonons (Eagles 1994a) or plasmons (Eagles 1994b), and the binding energy of pairs needs to be at least about 0.1 eV for pairs not to break up at room temperature, or larger if the break up does not occur until higher temperatures, as the higher T_c's inferred from measurements with *dc* (Enikolopyan *et al.* 1989) or pulsed-current measurements (Demicheva *et al.* 1990) imply. Thus the coupling with the high-frequency modes may be moderately strong, and so the single-particle masses and pair masses can be appreciably larger than m_e and $2m_e$ respectively. The radius of a polaron due to interaction with high-frequency modes of frequency ω will be of the order of $(\hbar/2m_b\omega)^{\frac{1}{2}}$, where m_b is the bare electron mass. For $m_b \approx m_e$ this radius is about 0.3 nm for $\hbar\omega \approx 0.37$-0.39 eV intramolecular phonons (McDonald and Ward 1961) or smaller for the larger $\hbar\omega$ expected for plasmons (Eagles 1994b), but we expect that the two polarons of a bipolaron will be separated by a larger amount because the Coulomb repulsion between the two polarons will keep them from coming too close. Hence the pair mass m_p may be close to twice the single-particle mass, as implied by the use of the Cooper-pair expression for the dispersion.

Note that the radius of a single polaron within a string and the pairing within a string due to interaction via high-frequency modes arise from different processes from those which cause the formation of the string itself. This is analogous to theories of stripe formation in high-T_c
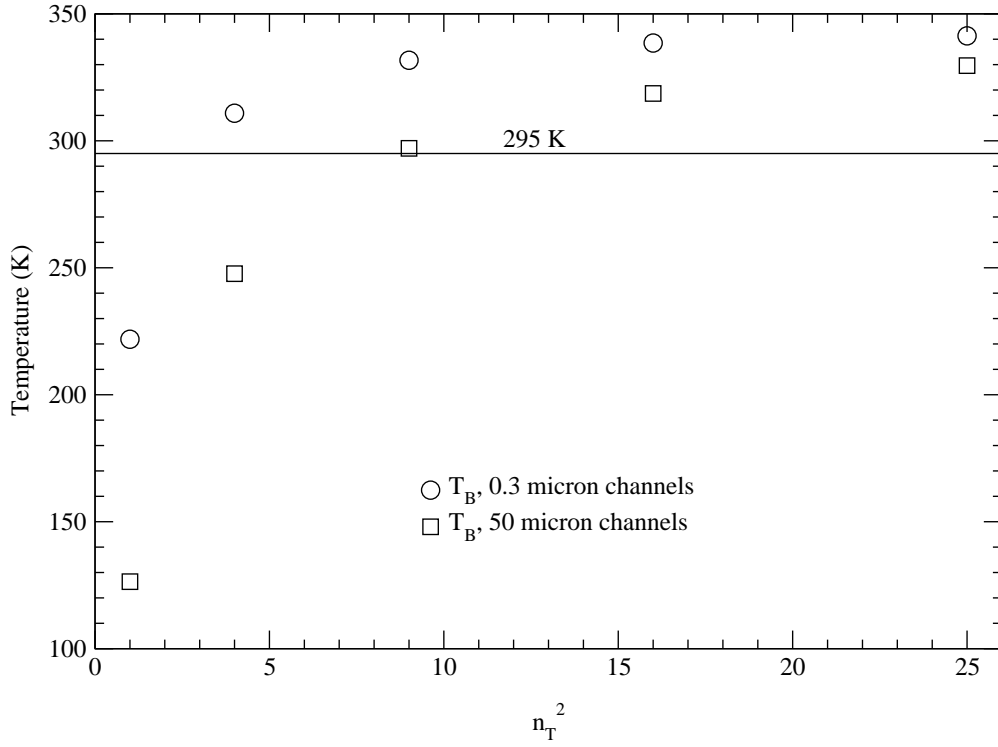
Figure 2: Condensation temperatures T_B for two values of L as a function of n_T^2, with the assumptions $a_T = 2$ nm, $s_T = 2^{\frac{1}{2}}$ at $T = 0$, $M_L = 3.3m_e$, $c = 0.5 \times 10^7 \text{cm}^{-1}$, and $p = 0.12$. At room temperature channels with $n_T^2 \leq 2$ are normal for $L = 0.3\mu\text{m}$ and channels with $n_T^2 \leq 8$ are normal for $L = 50\mu\text{m}$.

cuprates, where the interactions which cause the stripes to form are not generally thought to be the same as those which can cause pairing of carriers within a stripe. It is also worth noting that the concept of superpolaron strings in polymers was introduced many years before the concept of stripes in cuprates was considered.

Use of Eq.(7) implies that both spin states of the electrons are equally populated. Grigorov *et al.* (1990) have argued that the superpolaron strings will be ferromagnetic. However, we assume here that the larger pairing energies for singlet pairing will overcome the tendency of the superpolaron to be unstable when exchange energy is not minimised by a ferromagnetic arrangement.

For $m_s = 1.65 m_e$ and $k_F = 0.5\pi \times 10^7$ cm^{-1}, the Fermi energy $E_F = 0.057$ eV, and so the reduction factor F of the linear terms in the Cooper pair dispersion at room temperature is 0.56.

§5. A POSSIBLE INCREASE OF CONDENSATION TEMPERATURE WHEN CURRENT FLOWS

Figure 2 and calculations for larger n_T imply that the limiting value of T_B (for large n_T) is not greater than about 350K for the thicker films of figure 1. Some films called Type 1 (Enikolopyan *et al.* 1989) for which oxidation occurred slowly over several years without UV irradiation did have T_c close to room temperature. Other films, Type 2, for which UV irradiation was used for oxidation, had $T_c > 429$ K when 0.5 A currents were passed through a channel (Enikolopyan *et al.* 1989), and probably greater than 700 K when large pulsed currents were applied (Demicheva *et al.* 1990). The thicker films of figure 1 were of Type 2, and so might be expected to have T_c appreciably larger than room temperature when currents are passed through channels. We discuss a possible mechanism by which T_c can increase when current is applied. The proposed mechanism depends on the attractive force between filaments due to current-current interaction when currents are passing through. Because of the softness of the material, such forces can be expected to decrease the distance between filaments, and hence decrease the transverse masses of the channels. This gives rise to an increase in T_B. However, when we put in figures for a channel of width of the order of 1 μm, the rise in T_B is much too low to explain the data. Thus, in order to keep superconductivity at a given temperature when a current is applied, we postulate that the current concentrates in a smaller-width subchannel for which the current raises T_B above the relevant temperature. This will be energetically favourable because dissipation is avoided. We show below that, when $a_T = 2$ nm, $M_L = 3.3 m_e$, $(M_L/M_T) = 0.12$, and $c = 0.5 \times 10^7$ cm^{-1}, concentration of a 0.5 A current in a subchannel of initial lateral dimensions of about 0.24 μm is probably sufficient to raise T_B of the subchannel to above 429 K.

For an applied current of I in a square subchannel with n_T^2 filaments, the current per filament, I_f, is I/n_T^2. The pressure on any plane of filaments not at the surface of the array does not arise just from an external pressure at the surface, but is brought about by the forces on all planes in the array further out from the center than the plane being considered. Then, for odd n_T, for a plane m lattice constants from the center of the array, neglecting end effects, the pressure corresponds to an effective force per unit length on a filament at the center of the plane $F_{eff}(m)$ given by

$$F_{eff}(m) = (2I^2/a_T n_T^4) \sum_{i=m}^{q} \sum_{j=-q}^{q} \sum_{l=-q}^{q} [(j-i)/[(j-i)^2 + l^2)], \tag{13}$$

where

$$q = 0.5(n_T - 1), \tag{14}$$

and the triple sum excludes any terms with both $(j - i) = 0$ and $l = 0$. Then the average pressure over the whole array P_{av} is given by the average of $F_{eff}(m)/a_T$ i.e. by

$$P_{av} = [1/(q + 0.5)](1/a_T)[0.5F_{eff}(0) + \sum_{m=1}^{q} F_{eff}(m)]. \tag{15}$$

Using this result and taking a channel with $a_T = 2$ nm, and $D = 0.242\mu$m, corresponding to $n_T = 121$, we find, for an applied current of 0.5 A (=0.05 emu), that the average pressure on filaments at the centers of planes of the array is 2.4×10^6 dynes/cm^2. For OAPP films discussed by Grigorov *et al.* (1996), a pressure of 0.032 MPa gives rise to a reduction in thickness of about 3%, i.e. the compressive elastic modulus is about 1 MPa = 10^7 dynes/cm^2. At higher pressures there is also a slow plastic flow, and so calculations assuming the above elastic modulus will give a lower limit to the reduction in thickness caused. Thus we can expect the average stress due to current-current interactions to produce at least about a 24% average change in the filament separation a_T. A 24% decrease is equivalent to a decrease by a factor of exp(-0.27). For a small overlap of wave functions between channels, this percentage decrease in a_T will translate into a larger percentage increase of transverse bandwidth, by a factor of exp(f×0.27), where perhaps f lies between 3 and 5. If $f = 4$, then the ratio $p = M_T/M_L$ increases by a factor of about 2.9. From our computer program to solve Eq.(5), keeping other parameters as before, using $L = 20\mu$m [the thickness of the particular film used was not stated in Enikolopyan *et al.* (1989), but the results are not very sensitive to the thickness], decreasing M_T so that M_L/M_T increases by a factor of 2.9 from 0.12 to 0.35 for fixed M_L increases T_B for the largest value for n_T for which we have made calculations ($n_T = 70$) to above 429 K, as determined by experiments with 0.5 A currents passing through a channel.

To calculate a drift velocity for a given current for a reduced a_T, the carrier density goes up by the same factor as the current density increases. Thus we can calculate the drift velocity for the original values of a_T. A current of 0.5 A passing though a subchannel of cross section $(0.242 \ \mu$m$)^2$ corresponds to a current density within the subchannel of 8.54×10^8 A cm^{-2}, smaller than the order of magnitude estimate for the critical current density of 5×10^9 A cm^{-2} made from pulsed-current measurements by Demicheva *et al.* (1990). For a boson density of $ca_T^{-2} = 1.25 \times 10^{20}$cm^{-3}, the drift velocity corresponding to a current density of 8.54×10^8 A cm^{-2} is 2.13×10^7cm s^{-1}.

For a single-particle mass $m_s = 1.65m_e$ and a Fermi wave vector $k_F = 0.5\pi \times 10^7$cm^{-1}, the linear term in the boson dispersion corresponds to a velocity of $\hbar k_F/m_s = 1.10 \times 10^7$ cm s^{-1}, and thus the drift velocity corresponding to the quadratic term in the dispersion must be 1.03×10^7cm s^{-1}. For $M_L = 3.3m_e$, we deduce that the quadratic term in the energy is 0.10 eV, that the wave vector K corresponding to the drift velocity is 2.9×10^7 cm^{-1}, and that the linear term in the energy is 0.21 eV. Thus the total pair binding energy is larger than 0.31 eV. The bipolaron masses we deduce are quite sensitive to the boson concentration, e.g. if we assume a concentration twice as large as used above, the longitudinal boson mass deduced is increased to above $10m_e$. For a given mass, the condensation temperature increases faster than linearly with carrier concentration, partly because of the decreasing effect of the factor F of Eq.(12) as the concentration increases. Hence the mass to give a given condensation temperature at a fixed concentration increases faster than linearly with carrier concentration.

Note that Zhao (2003) has inferred energy gaps 2Δ of between 0.109 and 0.21 eV from tunnelling and Raman data in various carbon nanotubes, for which he infers from many types of

data that superconducting T_c's are sometimes greater than 600 K. Our inferred lower limit for the pair binding energy in OAPP is somewhat larger than thought to exist in carbon nanotubes. However, the masses we find are larger in OAPP, and so a larger binding energy and a further penetration into the Bose gas regime may occur in OAPP.

We consider that we have given a plausibility argument for a significant increase in superconducting T_c when a 0.5 A current passes. The case of much larger pulsed currents (Demicheva *et al.* 1990) is more complicated, and will not be discussed in detail here. However we make a few remarks about this case. First it was argued in Eagles (1994b) that the critical currents of about 60 A observed by Demicheva *et al.* (1990) may have been the combined total currents of several channels. Nevertheless, if e.g. we assume a a 20 A current in a channel of cross section $(1.002 \mu m)^2$ ($n_T = 501$), using Eq. (13) and arguments similar to those for the case of 0.5 A current, we find that the stress is larger by a factor of $40^2 \times 0.058 = 93$ than for the current concentrated in a subchannel of diameter 0.242 μm. For such large stresses, strong non-linear effects in the stress-strain curve will occur. We expect that when the stress is large enough to bring atoms sufficiently close to each other, then the elastic constants will increase to values similar to those of more normal solids, i.e. by a factor of the order of 10^6. Thus there will be a saturation of the strain, but it would be difficult to make any quantitative estimates.

Two other complications in the pulsed current case are: (i) When currents are close to those required to destroy superconductivity, there will be a competition between the increase in T_B due to increase of the transverse bandwidth by compressive stresses, and a lowering of T_B due to approach to the depairing current [cf. Kunchur *et al.* (2003)]. (ii) Another complication is the question of time scales for increases and decreases of a_T which will play a part in repetitive pulsed-current measurements.

§6. DISCUSSION

There are several arbitrary parameters in our theory besides the assumptions about the boson dispersion, and only three pieces of quantitative information to fit, viz. the approximate ratio of the lowest normal-state resistance of channels in films of two thicknesses, plus minimum values of room temperature and 429 K for superconducting transition temperatures at low currents and with 0.5 A currents flowing. Thus the best that can be said is that we have given a possible explanation for the results.

The $E(K)$ curves we have used for bosons in the longitudinal direction are based on results for Cooper pairs with an instantaneous electron-electron attraction. In that model the mass of the pair is twice that of the single particle whatever the strength of the electron-electron attraction. In bipolaron theories on the other hand, there are indications [see e.g. Macridin *et al.* (2004)] that the pair mass is only approximately equal to that of the single particles if the bipolaron binding energy is very small. However, we think that such a result could be modified when two different frequencies of excitations mediating the electron-electron attraction are involved. Then a small interaction mediated by a very high-frequency electronic excitation can affect the bipolaron binding energy appreciably while only giving a small change to the effective mass.

Our chosen $E(K)$ curves for the transverse direction and the temperature dependence of these curves for both directions are guesses. Although our chosen value for the value of the coefficient of the linear term for the transverse direction could be far from correct, if we choose another value for this coefficient which is not too small, then fairly similar results T_B can be obtained by adjusting the transverse mass to compensate. As an example, if we change s_T at $T = 0$ from $2^{\frac{1}{2}}$ to $0.5 \times 2^{\frac{1}{2}}$ and increase p from 0.12 to 0.155, T_B for n_T between two and ten

for both the shorter and longer channels is changed on average by well under 1%.

Another point which we mention is the following. The minimum number of nanofilaments to support superconductivity at room temperature deduced in §4 (nine for the longer channels) is less than the 30 deduced from a model for the metamagnetism given in Eagles (1998a). It is possible to increase the numbers of filaments required for superconductivity at room temperature in the model of the present paper by either increasing M_L or increasing M_T, but then a larger fractional decrease of M_T when 0.5 A currents are flowing is required to increase T_B to above 429 K. As an example, if we leave M_L as before but increase M_T to make $p = 0.062$ at low currents, then the largest values of n_T^2 for which the channels are normal at 295 K are $n_T^2 = 8$ for 0.3μm channels and $n_T^2 = 25$ for 50μm channels. This gives a ratio of cross sections of 3.1, not far from the mean of values of about five inferred from figure 1 and of about two from Enikolopyan *et al.* (1989), and a value of n_T^2 larger than 25 is needed to obtain superconductivity in the longer channels, in agreement with that of 30 required in the model of Eagles (1998a). If we now suppose that the 0.5 A current concentrates in a subchannel of width 0.202μm, corresponding to $n_T = 101$, then the decrease of a_T caused by the current is calculated to be by a factor of $0.66 = \exp(-0.42)$. If we assume that this corresponds to an increase of p by a factor $\exp(0.42\gamma)$, with γ taken as 4.2, then this gives an increase of p from 0.06 to 0.35, sufficient to increase T_B to above 429 K for large n_T. However the pair binding energy deduced from the larger drift velocities caused by concentrating the currents in a subchannel of smaller diameter than before now has to be larger than 0.76 eV, assuming the $E(K)$ curve for pairs retains the same form up to such rather large energies. This need not necessarily be the case. For the Cooper-pair problem with a finite-range interaction in two dimensions, it has been shown that, for large pair binding energies, a secondary minimum in $E(K)$ can develop, with a larger quadratic contribution to the energy for departures of K from its value at the minimum, (corresponding to a smaller mass at this secondary minimum) compared with that at $K = 0$ (Adhikari *et al.* 2000).

As an alternative to concentrating the current in a channel of smaller-diameter than before, we could consider the possibility that carrier concentrations in the channels may be increased by injection from the contacts when a current is applied, but we do not have a quantitative theory for this. A decrease of separation in distances between channels as current is increased was inferred from Josephson effect measurements at low temperatures in films of poly(phthalidylidenebiphenylene) (Ionov *et al.* 2002), but this does not give information about carrier concentrations within the channels. Also Skaldin (1991) analysed results on channels through thin films of polydiphenylenephthalide (Skaldin *et al.* 1990) to infer that cross sections of channels increase in proportion to applied currents, and the conductance of the channels increases in proportion to the cross sections, which implies that there is no increase in carrier concentration in the channels in this material.

The model of Eagles (1998a) involved quite a lot of assumptions, including that of spontaneous high currents circulating both within linear channels and round channels which form closed loops. This second type of assumption was not required in a recent interpretation (Eagles 2002) of large diamagnetism at low fields in films of oxidised atactic polypropylene (Rogachev and Grigorov 2000), and it is possible that the first type of assumption may not be necessary to explain the metamagnetism. Thus it would not be very surprising if the model of Eagles (1998a) did not give correct results for minimum numbers of nanofilaments required to support superconductivity. However, if we were to suppose that the model described there for metamagnetism is qualitatively correct, then we would expect that spontaneous currents would also flow round channels which form closed loops, and so a different explanation from that given in Eagles (2002) would be required to explain details of the magnetic susceptibility versus field curves in two samples of films of OAPP analysed in that paper.

More experimental results of the type shown in figure 1 would be of great interest, especially if they could be combined with measurements of the diameters both of the channels of medium resistance and of the superconducting channels. Many other types of experiments which might be worthwhile to perform on films of oxidised atactic polypropylene and other polymers are suggested in §3 of Eagles (1998a).

§7. MORE GENERAL DEPENDENCE OF T_B ON PARAMETERS

It is hoped that a paper with coauthors giving a more general discussion of how T_B depends on various parameters will be written later. However, to give some ideas about this in the meantime, we have done various calculations with all except one parameter fixed at values given in the caption to figure 2, while one is varied, for several values of n_T. A typical value of channel length $L = 20\mu$m is considered if L is not the parameter to be varied. We find:

1. T_B is proportional to the reciprocal of the longitudinal mass. However, bipolaron binding energies will decrease as masses decrease, making it more difficult to get on the Bose gas side of the BCS-Bose gas transition for small masses.

2. T_B also increases as the transverse mass decreases but not so fast. T_B is approximately proportional to $M_T^{-0.184}$ for values of M_T/M_L that lie between 0.04 and 0.3 for $L = 20\mu$m, $n_T = 10$ and other parameters as in the caption to figure 2. However, when M_T becomes a significant fraction of M_L, then the quasi one dimensionality is lost, and so it may be less easy to obtain a large bipolaron binding energy. The increase in T_B as M_T decreases (subject to the limitation mentioned above) implies that T_B will increase rather faster as the transverse lattice constant a_T decreases, assuming $M_T \propto a_T^{-\nu}$, with ν in the range of 3 to 5.

3. T_B increases faster than linearly as the boson concentration increases. For $n_T = 10$, $L = 20\mu$m and other parameters as in the caption to figure 2, T_B increases with c for ca_T between 0.04 and 0.5 approximately as a power between 1.61 and 1.75, with the exponent rising slowly as c increases.

4. We have performed calculations for n_T from 1 to 10 for values of L from 0.1 to 1000 μm for other parameters as in the caption to figure 2. For $n_T = 1$, T_B decreases monotonically with increasing L and decreases by 62% as L increases by four orders of magnitude. For other values of n_T, T_B goes through a maximum as L begins to increase, and then decreases, with the rate of decrease slower for larger n_T, e.g. as L changes from 100 to 1000 μm, T_B decreases by 4.0% for $n_T = 5$ and by 2.9% for $n_T = 10$. These slow decreases do not appear to be sufficient to account for the fact that room-temperature superconductivity has never been reported through films of thickness greater than 100μm (Enikolopyan *et al.* 1989). This is more likely to be associated with the difficulty of forming stable long and straight conducting channels (Grigorov 1992).

5. As the zero temperature value of the ratio of linear to quadratic contributions to the transverse bandwidth is varied from 0 to 3, for $n_T = 10$, $L = 20\mu$m and other parameters as in the caption for figure 2, T_B increases by 24%.

6. Figure 2 gives examples of how T_B depends on n_T. Compared with the case of pure quadratic dispersion discussed by Eagles (1998b), the initial rate of rise of T_B with n_T is much faster for the combined linear plus quadratic dispersion.

§8. WHAT IS NEEDED FOR HIGH T_c BESIDES QUASI ONE DIMENSIONALITY?

We have suggested that electron-phonon interactions and/or electron-plasmon interactions are the cause of the pairing of electrons in channels through OAPP. However, other polar quasi one-dimensional materials can be expected to have electron-phonon interactions of comparable strength, and, for appropriate current carrier concentrations, we might at a first guess think that electron-plasmon interactions could comparable magnitude to those in channels through OAPP. So why have not many other types of quasi one-dimensional materials shown superconductivity at very high temperatures? We do not have a definite answer to this question, but make a few

tentative suggestions here.

First we note that the only other two quasi one-dimensional materials that we are aware of for which room-temperature superconductivity has been claimed besides those discussed in this paper are, (i) carbon nanotubes (Tsebro *et al.* 1999, Zhao and Wang 2001, Zhao 2003), and (ii) powdered mixtures of $PbCO_3.2PbO + Ag_2O$ (Djurek *et al.* 2001). The structure of the superconducting components of the system studied by Djurek *et al.* has been suggested to contain well separated Ag-O chains which are thought to be the main channels for possible superconductivity in this system. The only atoms in carbon nanotubes are carbons, and the linear chains thought to give the superconductivity in the system of Djurek *et al.* are composed of silver and oxygen atoms. In both of these systems the quasi one-dimensional conduction takes place by electrons which do not have a d-electron component. Further, for OAPP and other elastomers, according the the theory of Grigorov (1990,1991), nanofilaments from which the conducting channels are composed pass through spaces in the polymeric system, and so the electrons in the nanofilaments of the channels avoid all atoms, and do not involve d-electrons or any other atomic orbitals. Thus, on the basis of what types of materials are thought by some to show room-temperature superconductivity, we make our first hypothesis that, besides quasi one-dimensional conduction, the conduction electrons must not have a d-electron component. If d electrons take part in the conduction, Coulomb repulsion can be expected to be large, and so, for given strengths of electron-phonon and electron-plasmon interactions, it will be more difficult to obtain large pair binding energies, assuming s-pairing symmetry. (We do not discuss d-wave superconductivity here, for which on-site Coulomb repulsion does not cause a problem).

However, the above criterion does not tell the whole story, because there are quasi one-dimensional polar organic materials which do not involve d-electrons but which only show superconductivity at low temperatures, or instead form charge-density waves. The squares of matrix elements of electron-plasmon interactions are proportional to $\epsilon_h^{-3/2}$, where ϵ_h is the high-frequency dielectric constant of the material (see e.g. Bose and Gayen, 2004) and the squares of the matrix elements for Fröhlich interactions interactions with longitudinal optical phonons are proportional to $(1/\epsilon_h - 1/\epsilon_s)$ (Fröhlich 1954), where ϵ_s is the static dielectric constant. Thus a low high-frequency dielectric constant is important for strong interactions mediated via plasmons, and for interactions mediated by longitudinal optical phonons a low ϵ_h and a fairly high ϵ_s are necessary. The high-frequency dielectric constant of polypropylene is $1.57^2 \approx 2.2$ (Brandup and Immergut 1989), and an effective dielectric constant for graphite has been reported to be 1.4 (Egger and Gogolin 1998). The value of 2.2 for polypropylene is not much smaller than the value of ϵ_h for several organic conductors and superconductors (Jérome and Schultz 1982). Thus, other things being equal, electron-plasmon interactions might be expected to be of similar magnitude in OAPP and some quasi one-dimensional organic superconductors such as the $(TMTSF)_2X$ series. The static dielectric constant ϵ_s is quite high in films of OAPP having superconducting channels. The increase of ϵ_s during heat and ultra-violet treatment is associated with an increase of rotatable dipolar groups. However, when the concentration of dipolar groups becomes sufficiently large, more atoms become ionised (Grigorov 1990), and so at least part of the increase in ϵ_s should help to increase the polarisation associated with lattice vibrations, and hence to increase interactions between electrons and longitudinal optical phonons. We also note that some of the intramolecular vibrations in polypropylene have very high frequencies (McDonald and Ward 1961), and so contributions to bipolaron binding energies from interactions with these phonons will be relatively large for a given strength of electron-phonon interation. We have not found information on ϵ_s in the $(TMTSF)_2X$ family yet, but the charges on the whole molecules involved are or of magnitude half or one electronic charge, and so the average charge density over the large (TMTSF) molecules is expected to be small. Thus the polarisation density from their

motion and hence ϵ_s may not be very large. Hence electron-electron interactions mediated by longitudinal optical phonons may be significantly larger in OAPP than in (TMTSF)$_2$X. Another factor that may reduce pairing energies is the probable non-negligible on-site Coulomb repulsion for p-electrons in the (TMTSF)$_2$X materials, and the possible interference of repulsive interactions via magnetic excitations with attraction via phonons and plasmons. Although OAPP has some unusual magnetic properties besides fairly large diamagnetism at low fields (Smirnova et al., 1998, Enikolopyan *et al.* 1989, Grigorov *et al.* 1996, Rogachev and Grigorov 2000), we are not aware of any magnetic excitations in this material which might mediate an electron-electron repulsion. Another factor to take into account is competition between superconducting pairing and instabilities associated with charge- or spin-density waves, which are more likely to be of importance for electrons in a tight-binding type of band structure rather than for the nearly-free bare electrons which we think occur in the conducting channels through films of OAPP. A final property required for high condensation temperatures is a fairly low bare mass, so that bipolaron masses need not necessarily be excessively large.

So, based on arguments given in the last two paragraphs, we tentatively suggest eight properties which are helpful for very high-temperature s-wave superconductivity. These are:
1. Quasi one-dimensional conducting channels. The restricted dimensions help bipolaron formation. Mourachkine (2004) is also of the opinion that quasi-one dimensionality is a requirement for room-temperature superconductivity, but suggests that bisolitons are probably the type of pairs involved. This may well be the case in some materials.
2. In order to keep Coulomb repulsion low, conduction electrons in these channels should not have any d-orbital component, and should preferably be electrons which avoid atoms altogether as in channels in OAPP.
3. The quasi one-dimensional channels should lie in a background material with a low ϵ_h in order to help to give large electron-plasmon interactions.
4. The background material should also have a high ϵ_s, so that interactions with longitudinal optical phonons can be large. The extent to which the increase in ϵ_s during heat and ultra-violet treatment of atactic polypropylene causes increases in electron-phonon interactions is not known at present. This is because the increase in ϵ_s is associated with increase of rotatable dipolar groups, but also with increases in numbers of ionised atoms.
5. The presence of very high-frequency phonons is an advantage, as this permits larger bipolaron binding energies for a given strength of electron-phonon interaction.
6. There should be a lack of competition with other forms of order than superconductivity, such as charge- or spin-density waves. Such competition is less likely to occur if the bare electrons in the channels are closer to being free-electron like than to being describable by tight-binding types of band structure.
7. There should be a lack of competition between repulsive interactions mediated via magnetic excitations and attractive interactions mediated by plasmons and phonons.
8. A fairly low bare-electron mass in the channels is required in order to give a chance for the Bose-Einstein condensation temperature of the pairs to be large.

OAPP has at least seven of these properties, and possibly all eight, although number 7 above is uncertain. Carbon nanotubes do not satisfy property 4, but this is probably compensated by the unusually low effective ϵ_h, making electron-plasmon interactions unusually strong. We do not have enough information about the compound studied by Djurek et al. (2001) to know how many of the above eight properties are satisfied for this compound apart from the first two.

§9. CONCLUSIONS

We have given a plausible explanation for the fact that the minimum resistance of normal channels through oxidised atactic polyproplene (OAPP) films increases with film thickness more slowly than linearly. The explanation is based on calculations of the condensation temperature T_B of bosons in arrays of filaments when the bosons have a mixed linear plus quadratic dispersion, as indicated by published results for the Cooper-pair problem. The condensation-temperature calculations show that the minimum cross section of arrays which can support superconductivity at a given temperature increases as the length of the array increases. To explain the observations in detail with the model, we need to assume that, even for large tranverse dimensions of the filament arrays, the superconducting T_c at low currents is not more than about 350 K. To explain higher T_c's inferred from data involving higher applied currents, we need to postulate that T_c increases as the current increases, at least for not too large currents. A possible reason for this is shown to be related to the fact that current-current interactions produce compressive stresses which cause significant compression of the array in the elastomer, atactic polypropylene, and that this gives rise to larger transverse bandwidths with an associated increase in T_B. However, in order to explain the results quantitatively, we need to suppose that the applied current concentrates in a subchannel of lateral dimensions of about 0.24μm, smaller than the probable diameters of the whole channels, of the order of 1 μm (Demicheva et $al.$ 1990).

APPENDIX

It would be difficult to do a proper calculation of the parameter s_T of equation (1). In this Appendix we present two types of calculations which probably give upper and lower limits for s_T.

For a simple anisotropic system with no cut offs in wave vectors, we guess that the ratio r of coefficients of linear and quadratic terms for transverse motion would be given by

$$r = 2k_{FT} = 2k_{FL}(m_T/m_L)^{\frac{1}{2}}, \qquad (A1)$$

where m_L and m_T are the longitudinal and transverse masses for single particles. The factor of two in equation (A1) instead of four given in equation (6) arises because of the three dimensionality of the system. Equation (A1) implies

$$s_T = 2p^{-\frac{1}{2}}(k_{FL}/k_{MT}), \qquad (A2)$$

provided $m_L/m_T = M_L/M_T = p$. Here $k_{MT} = \pi/a_T$.

However, when k_{MT} is small compared with k_{FT} given above, then, for most values of the longitudinal component of the single-fermion wave vector, the transverse wave vector is limited by the zone edge value k_{MT}. The fraction f of longitudinal wave vectors for which the transverse wave vector at the Fermi surface is less than k_{MT} is given by

$$f = 1 - (1 - E_{MT}/E_F)^{\frac{1}{2}}, \qquad (A3)$$

where $E_{MT} = (\hbar^2/2m_T)k_{MT}^2$. If E_{MT}/E_F is fairly small, then we can approximate f as

$$f \approx (1/2)(E_{MT}/E_F) = (p/2)(k_{MT}/k_{FL})^2, \qquad (A4)$$

if $m_L/m_T = M_L/M_T = p$. For those longitudinal wave vectors for which the transverse wave vector is limited by the zone edge, we do not expect any contribution to the linear term in the Cooper pair dispersion provided that any gap at the zone edge is large compared with the pair

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binding energy. In that case we infer from equations (A2) and (A4) that s_T, the ratio of linear and quadratic contributions to the transverse pair bandwidth is given by

$$s_T \approx p^{\frac{1}{2}}(k_{MT}/k_{FL}) \qquad \text{(A5)}$$

The assumption that the excitations from the parts of the Brillouin zone where the transverse component of the Fermi wave vector extends to the zone edge do not contribute to the linear terms in the Cooper pair dispersion depends on any gap at the zone edge being large compared with the pair binding energy. In the opposite limit when the pair binding energy is large compared with such gaps, we expect that such gaps do not have much effect. So, in that case s_T might be given by equation (A2). Thus equation (A2) probably gives an upper limit for s_T and equation (A5) a lower limit. Since we do not which of the two limits is a better approximation, we use the geometric mean of the two, which turns out to have the simple value

$$s_T = 2^{\frac{1}{2}}. \qquad \text{(A6)}$$

This is the result we assume for $T = 0$. Our numerical calculations indicate that, if we use different values of s_T which are not very small, then changes in s_T can be approximately compensated for by appropriate changes in p.

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